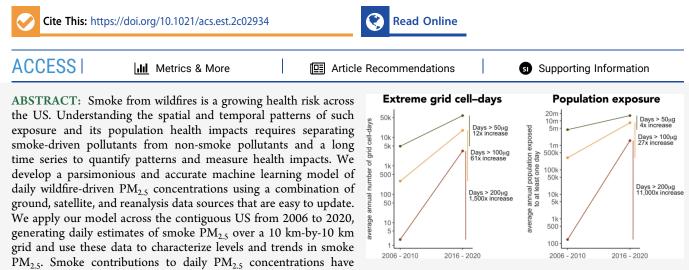


Daily Local-Level Estimates of Ambient Wildfire Smoke $\text{PM}_{\rm 2.5}$ for the Contiguous US

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increased by up to 5 μ g/m³ in the Western US over the last decade, reversing decades of policy-driven improvements in overall air quality, with concentrations growing fastest for higher income populations and predominantly Hispanic populations. The number of people in locations with at least 1 day of smoke PM_{2.5} above 100 μ g/m³ per year has increased 27-fold over the last decade, including nearly 25 million people in 2020 alone. Our data set can bolster efforts to comprehensively understand the drivers and societal impacts of trends and extremes in wildfire smoke.

KEYWORDS: particulate matter, wildfires, smoke, aerosols, machine learning

INTRODUCTION

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Exposure to ambient fine particulate matter (PM_{2.5}) has long been recognized as a leading environmental determinant of health outcomes and is estimated to cause millions of premature deaths globally and 48,000 deaths in the US every year.¹ Efforts to reduce this health burden require understanding the main sources of PM2.5 and how pollutant concentrations from these sources are distributed across human populations. While many countries have made remarkable strides in reducing key anthropogenic emission sources and thus PM_{2.5} concentrations, albeit sometimes from very high levels,^{2,3} other PM_{2.5} sources appear to be growing rapidly. Of key concern in many parts of the world is the growing contribution of wildfire smoke to pollution concentrations.⁴⁻⁹ Increases in wildfire smoke are being driven in a substantial part by a warming climate, which increases the flammability of fuels and in turn the occurrence of larger, more extreme wildfires which emit more smoke.¹⁰⁻¹² Lofted high into the atmosphere, resulting pollutants can travel long distances from their source fire, potentially affecting ground pollution levels and health outcomes thousands of kilometers away.¹³ Because smoke from wildfires can also generate abrupt extreme pollutant concentrations and because emitted pollutants might differ in their toxicity relative to analogous

non-wildfire species,¹⁴ accurate measurement of the location and level of smoke-attributable surface pollutant concentrations is key for understanding the societal impacts of growing wildfire risk.

However, accurate attribution and measurement remain empirically challenging. Ground monitors measure concentrations of total pollutants and cannot be easily used to attribute pollutants to their sources without additional information. Atmospheric chemical transport models (CTMs) offer one common approach to linking wildfire activity to concentrations of key pollutants and downstream human impacts,^{15–20} but their use faces a number of difficulties.²¹ First, uncertainties in wildfire emission inventories have been shown to lead to substantial differences in CTM-estimated smoke PM_{2.5} concentrations, with up to 20fold differences in estimated regional wildfire-attributed PM_{2.5} concentrations²² when different leading inventories are fed

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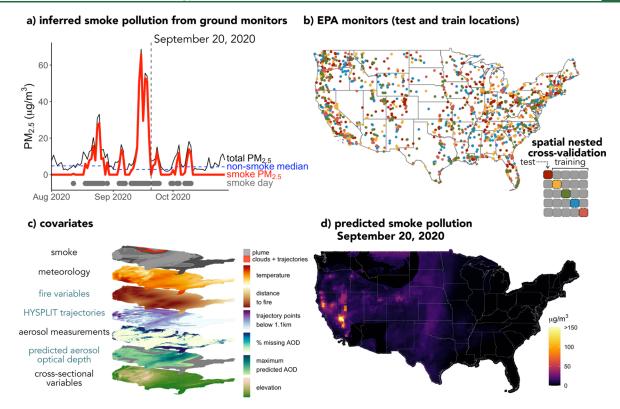


Figure 1. Model predictions are derived from inferred smoke pollution at ground monitors and remotely sensed covariates. (a) Total fine particulate matter ($PM_{2.5}$, black line) is observed at ground monitors. Plumes and trajectories from fires are used to identify days with smoke (gray points). Non-smoke median $PM_{2.5}$ (blue dashed line) is the station- and month-specific median of $PM_{2.5}$ on non-smoke days, and smoke $PM_{2.5}$ (red line) is total $PM_{2.5}$ above the non-smoke median on days with smoke overhead. (b) Smoke $PM_{2.5}$ is inferred at EPA monitor stations (points) throughout the US, which are then grouped into five disjoint folds for model training and validation to perform spatial nested cross-validation (inset). (c) Smoke days (top layer) are defined based on the presence of smoke plumes (light gray areas) or the presence of 50 HYSPLIT trajectory points below 1.1 km and AOD missingness over 75% (red areas), as a proxy for cloud cover that would prevent the identification of smoke plumes. Other covariates included in the model are used to predict smoke $PM_{2.5}$ on smoke days and include both time-varying and cross-sectional variables. Covariates labeled in black are directly measured by other products, while those in blue are derived. (d) Resulting predictions are a 10 km resolution gridded product of daily smoke $PM_{2.5}$ over the contiguous U.S. from 2006 to 2020. All time-varying covariates and predictions shown are from September 20, 2020. State outlines are reproduced with permission from the US Census Bureau TIGER/Line Shapefiles.⁵¹

into the same transport model, although recent efforts have shown that satellite-based emission calibration can improve the simulation accuracy.²³ Second, the complex chemistry regimes during wildfire (in particular, production of organic aerosols) and hyperlocalized meteorology are often not well-captured by CTMs, leading to concerns over their ability to correctly model the transport of wildfire smoke.^{24–26} Third, parameterized heights of plume injections and the vertical allocations of emissions differ considerably across different CTMs and often compare poorly with observed injection heights, leading to discrepancies in surface $PM_{2.5}$ concentrations when compared with in situ observations.^{27,28}

As a complement to CTM-based approaches, recent efforts have sought to use statistical approaches to directly estimate smoke $PM_{2.5}$ concentrations without explicitly parameterizing the underlying physical processes. The rapid expansion in the frequency and availability of satellite imagery has led to numerous satellite-based estimates of wildfire smoke movement,^{29,30} but commonly used satellite products (e.g., aerosol optical depth or satellite-derived plume boundary estimates) do not provide precise information on where in the atmospheric column the smoke occurs, making it difficult to use this information alone to infer surface concentrations. Instead, non-CTM-based approaches typically include the use of ground monitor data to infer wildfire-attributable pollutant concentrations. The main empirical challenges of non-CTMbased approaches for estimating wildfire-attributable air pollution are the accurate partitioning of smoke- and nonsmoke-sourced pollutants and the estimation of pollution concentrations in locations where ground monitors are not available. Studies have taken multiple approaches to solving the partitioning problem, including building statistical models of all the main sources of pollution and estimating the model with and without wildfire variables,⁴ estimating smoke PM_{2.5} at a given monitor location as anomalous deviations from estimated seasonal non-smoke background PM2.5 concentrations,^{5,31} using particle trajectory models to estimate wildfire-influenced and non-influenced surface pollution,³² and simply attributing extreme positive PM25 anomalies during summer and fall wildfire seasons in the US West to wildfires.⁶ Alternate approaches have also been used to estimate smoke pollutant concentrations for locations without monitors, including fitting statistical models that relate monitor observations to input variables and then using the model to estimate concentrations in non-monitor locations⁴ or interpolating monitor observations across space and inferring the smoke contribution using one of the abovementioned techniques.^{5,6,31} The primary goal of most of these analyses has been to shed light on the longer-term trends in smokederived $PM_{2.5}$ and how smoke contributions to total $PM_{2.5}$ have changed over space and time.

Here, we build on these non-CTM-based approaches to estimate daily, local-level wildfire-smoke-attributable surface PM_{2.5} across the contiguous US. We combine information from satellite-based smoke plume identification and simulations of air trajectories from fire locations to identify when smoke is in the air. Then, similar to other studies, we attribute anomalous deviations from background levels of PM2.5 to smoke when smoke is plausibly in the air. To estimate smoke PM2.5 at locations without monitors, we take advantage of the additional available information from satellite and reanalysis products and build a machine learning model to predict daily smoke PM_{2.5} over the contiguous US. This combines the strengths of non-CTM-based approaches, which carefully attribute observed ground PM_{2.5} concentrations to smoke, and satellite data which provide additional information in locations without monitors. In contrast to many past machine learning-based efforts predicting total PM2.5, our goal is to isolate smoke-driven changes in PM_{2.5} from other changes in PM_{2.5}. Our efforts are thus a complement to recent machine learning-based efforts at predicting variation in total PM2.5, for example, Di et al.³³ and Reid et al.³⁴

These smoke $PM_{2.5}$ predictions are designed with downstream applications and inference tasks in mind. We sought predictions that accurately captured local short-run variation in smoke $PM_{2.5}$ levels and extremes over sufficiently long time series and across a large spatial domain. Simultaneously, we sought a tractable model with inputs that would be easy to update in the future. These dual goals led us to produce daily predictions of smoke $PM_{2.5}$ at a 10 km resolution using a model with high predictive performance at both low and extreme levels of smoke $PM_{2.5}$, which we then used to understand the levels, trends, and changing daily extremes in smoke $PM_{2.5}$ concentrations across the contiguous US. We also connected these estimates to data on income, race, and ethnicity to understand differential patterns of exposure and how these patterns are evolving over time.

METHODS

Our approach has three main steps. First, we identify days when smoke was overhead ("smoke days") from satellite imagery-based plume classification and when clouds may be obscuring plumes, from simulated air trajectories originating at fires (Figure 1c, smoke). Second, we construct a ground-based measure of smoke PM2.5 by calculating PM2.5 anomalies at Environmental Protection Agency (EPA) monitoring stations (Figure 1a,b), measured as deviations from recent locationand month-specific median PM2.5 on non-smoke days and then attribute positive anomalies to smoke if our approach in the first step indicated that there was smoke overhead on that day. Third, because pollution monitoring stations are infrequent in space and typically have many missing observations, we train a model that predicts the station-based ground smoke PM2.5 estimated in the second step, based on a set of spatially and temporally consistent inputs (Figure 1c), validating the model on daily time series from held-out EPA stations (i.e., stations not used in model training) and on daily time series from an independent set of private monitors that were not used in model development. Finally, we use the model to produce daily 10 km gridded estimates of smoke pollution over the contiguous US (Figure 1d). Unless noted otherwise, analyses were performed using the R programming language.³⁵

Defining Smoke Days. To understand when smoke from fires may be affecting ground pollution levels, we construct a binary classification of smoke days for each cell of a 10 km grid covering the contiguous US based on two sources of information: hand-annotated smoke plumes from satellite imagery and modeled air particle trajectories from fire locations to aid in identifying smoke when clouds may obscure plume identification. First, we use data on smoke plumes from the National Oceanic and Atmospheric Administration (NOAA) Hazard Mapping System (HMS),³⁶ which are analyst-identified plume boundaries based on visible bands of satellite imagery.^{30,37,38} A grid cell is classified as a smoke day if it has any intersection with a smoke plume on a given day (Figure S2a,b). The first full year for which the HMS plume data are available is 2006, which limits the start date of our study period.

Second, given the potential for smoke plume boundaries to miss areas affected by smoke, especially when clouds hinder identification, we build on recent work 29,32 and simulate air packet trajectories from smoke-producing fire points detected by HMS using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model.^{29,38,39} Using these runs, we classify grid cell days as smoke days if they are missing more than 75% of 1 km aerosol optical depth observations (AOD, 0.47 μ m; based on a moderate resolution imaging spectroradiometer (MODIS) multi-angle implementation of atmospheric correction (MAIAC) daily observations) and have a count of at least 50 HYSPLIT trajectory points in the lowest height quintile (0-1.1 km above ground level) within a 50 km buffer of the location of interest. These thresholds are selected to yield a set of smoke days with anomalous PM2.5 values similar to the original set of days (Figure S2e,f; see the Supporting Information for more details).

Calculating Ground-Based Measures of Smoke PM_{2.5}. We then combine classification of smoke days with data on daily average $PM_{2.5}$ concentrations from 2019 EPA monitoring stations throughout the US to define daily time series of smoke $PM_{2.5}$ at each station.⁴⁰ We first define $PM_{2.5}$ anomalies as deviations from recent month- and location-specific median values on non-smoke days (Figure 1a)

$$\widetilde{PM}_{idmy} = PM_{idmy} - \overline{PM}_{imy}^{NS}$$
(1)

where PM_{idmy} is the $PM_{2.5}$ at station *i* on day *d* in month *m* and year *y* and \overline{PM}_{IMY}^{NS} is the 3 year location- and month-specific median $PM_{2.5}$ on non-smoke days. This median is calculated as

$$\overline{PM}_{IMY}^{NS} = \text{median}(\{PM_{idmy}|i = I, m = M, Y - 1 \le y \le Y + 1, \text{smoke}_{idmy} = 0\})$$
(2)

with smoke_{idmy} being a binary variable indicating smoke day classification. We use medians rather than means to prevent days with extreme $PM_{2.5}$ that are not smoke days from affecting the background $PM_{2.5}$ estimates, as is occasionally the case in our data. Furthermore, using 3 year medians, we allow the measure of background non-smoke $PM_{2.5}$ to change over time in each location to capture trends in non-smoke $PM_{2.5}$ over time, including potential declines in anthropogenic emissions. We then define ground-based smoke $PM_{2.5}$ as anomalies above the median on days in which smoke was overhead

$$smokePM_{idmy} = max(PM_{idmy} \times smoke_{idmy}, 0)$$
(3)

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Our approach to identifying smoke $\rm PM_{2.5}$ at monitoring stations is similar to other recent studies. 5,31 Similar to previous non-CTM efforts, these estimates rely on the HMS plume boundaries for identification of when smoke is affecting PM_{2.5}. We find that having a smoke plume overhead is associated with an average of 4.5 μ g/m³ increase in PM_{2.5} after controlling for monitor-specific averages and average differences in PM_{2.5} between states, months, and years (Table S1). We also find that in monitor time series, plumes align temporally with spikes in $PM_{2.5}$ (Figure S5). Furthermore, to counter omissions when clouds may obscure smoke plumes,²⁴ we included locations with potential cloud cover and air trajectories from known fires (see the "Defining Smoke Days" section mentioned above). Nevertheless, smoke days may be a conservative estimate of the locations with air quality impacted by smoke due to undetected plumes under cloud cover, during nighttime when satellite-based plume segmentations are unavailable, or where smoke is dilute and difficult to identify in satellite imagery.^{5,29} See more detailed discussion in the Supporting Information ("Plume accuracy").

To further confirm that our method of constructing smoke PM_{2.5} from ground station anomalies is indeed picking up PM_{2.5} from smoke and not from local time-varying sources of PM_{2.5} unrelated to smoke, we apply our method to harmonized speciated data from Interagency Monitoring of Protected Visual Environments (IMPROVE)⁴¹ and Chemical Speciation Network $(CSN)^{42}$ monitors.⁴³⁻⁴⁶ If our approach is identifying smoke-sourced PM2.5 and not PM2.5 from other sources, then species most likely to be present in smoke PM_{2.5}—which includes organic carbon throughout the US and perhaps additionally sulfates from agricultural fires in the southeastern US^{47,48}—will increase on smoke days but other non-fire-associated species will not increase. This is indeed what we find: the share of anomalous PM_{2.5} made up by organic carbon increases substantially, particularly on days that our method would predict are very high smoke days and particularly in settings where background sources of other $PM_{2.5}$ are low (such as in the western US) (Figure S6). In areas where background PM_{2.5} levels are higher and where agricultural burning is more common, such as in much of the southeastern US and in the southern Central Valley of CA, both organic carbon and SO₄ increase in importance on smoke days—although, as expected,²¹ their contribution to total PM_{2.5} on that day is still smaller than in Western areas where background PM_{2.5} is low. In both settings, non-wildfire sources of PM_{2.5} (such as dust and elemental carbon) do not increase on smoke days. These results provide confirmatory evidence that our method of assigning anomalous $\mathrm{PM}_{2.5}$ to smoke on days with smoke plumes overhead is indeed picking up wildfire-sourced $PM_{2.5}$ and not some other correlated $PM_{2.5}$ source.

Predicting Smoke PM_{2.5}. Because of the limited spatial and intermittent temporal coverage of ground-based PM_{2.5} monitors in the US, estimates of smokePM_{*idmy*} from eq 3 with time series for more than 5 years are only available at roughly 1400 ground locations. In addition, the average person in the US lives more than 20 km from the nearest pollution monitor.⁴⁹ To capture local short-run variation in smoke-PM_{*idmy*} over space and time, we therefore incorporate additional sources of information. One option is to use existing high-resolution gridded daily estimates of PM_{2.5}, from which anomalized estimates of PM_{*idmy*} could be constructed to

plug into eq 3. Numerous machine-learning-based efforts have succeeded in generating such estimates, but using them faces a number of challenges. First, many estimates are not updated after their initial publication and are available only through earlier years (e.g., Di et al.³³) and/or for a geographic region such as the Western U.S (e.g., Reid et al.³⁴), making them difficult to use to estimate up-to-date countrywide smoke pollution. Second, these $PM_{2.5}$ predictions are from a model trained to predict total $PM_{2.5}$, not smoke $PM_{2.5}$, and as such could potentially not be optimized for our particular task of interest. Finally, estimates could be hard to update, as highest performing model runs depend on a large number of inputs or outputs from other models that themselves are infrequently updated, making updating computationally burdensome.^{33,34}

A second option is to train a model to predict smokePM_{*idmy*} directly, using inputs from satellite, ground-measured, and reanalysis data sets that are straightforward to update. Because in this approach the model can be optimized to predict the outcome of interest—including focusing on inputs that predict smoke PM_{2.5} (e.g., distance to fire) rather than total PM_{2.5} (e.g., road density)—and can be carefully validated with downstream inference tasks in mind, we take this second option as our main approach.

Model Inputs. Building on earlier statistical-model-based smoke pollution prediction efforts,^{4,5} we extract or compute a set of model inputs that include meteorology, derived fire variables from HMS fire points, counts of HYSPLIT trajectory points, direct aerosol measurements, AOD predictions, and cross-sectional land use and elevation (Figure 1c and Table S3). Model inputs were merged to a consistent 10 km grid for smoke PM_{2.5} training and prediction. The grid is constructed to cover the contiguous US, based on US Census Bureau TIGER/Line county borders.^{50,51}

Detailed information on model inputs is provided in the Supporting Information. Briefly, meteorological inputs to the model include daily mean, minimum, and maximum of the planetary boundary layer, mean wind speed in the eastward and northward directions, mean air temperature and dewpoint temperature at 2 meters, total precipitation, and mean sea level and surface pressure, all drawn from European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis 5th Generation (ERA5) land or global products.^{52,53} Second, to capture the potential for characteristics of proximate fires to affect smoke PM_{2.5}, we used fire points from NOAA HMS³⁶ identified by trained analysts and used distance to these points as a model input. Third, counts of HYSPLIT trajectory points within a 50 km buffer by quintiles of height above the ground level were used as the input. Fourth, we used two complementary approaches for measuring aerosols: a reanalysis-based approach with consistent data but coarse spatial resolution, based on Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) aerosol optical thickness (AOT) data,⁵⁴ and a satellite-based approach with higher resolution but many missing observations from MODIS MAIAC AOD.55-57 Observations in the MAIAC product were first predicted using a gradient boosted tree model⁵⁸ with AOT anomalies on smoke days, meteorology, fire variables, elevation, and land cover as inputs (Table S2), and the resulting predicted AOD measures were used as the input to the smoke prediction model. Finally, we used time-invariant data on land use and elevation from USGS National Elevation Dataset⁵⁹ and National Land Cover Dataset⁶⁰ as additional inputs.

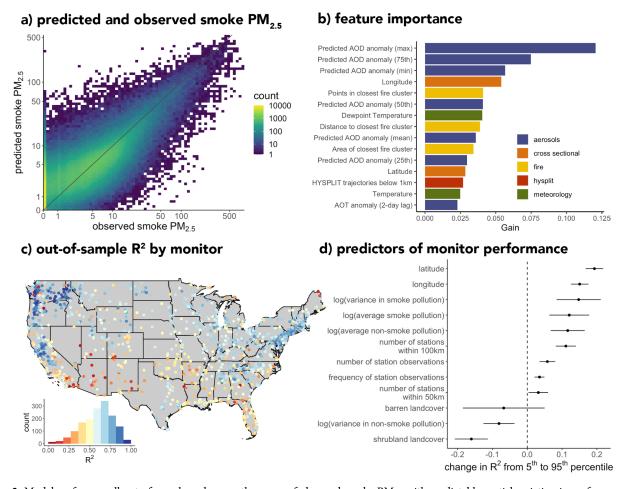


Figure 2. Model performs well out-of-sample and across the range of observed smoke $PM_{2.5}$ with predictable spatial variation in performance. (a) Color indicates count of monitor smoke days within bins of observed (horizontal axis) and predicted (vertical axis) smoke $PM_{2.5}$. Axes are pseudo-log-transformed, and color scale is log-transformed. The black line indicates the 1–1 line or where predictions perfectly match observations. (b) Feature importance for the top 15 features (vertical axis) from the final model, as measured by gain (horizontal axis) and colored by a broad category of the feature type, includes aerosols, cross-sectional information on elevation and land cover, fire variables, HYSPLIT, and meteorology. (c) For each monitor with at least 50 observations (points on map), R^2 is calculated over all smoke days with available smoke $PM_{2.5}$ using predictions from the model in which that station was out-of-sample. The inset histogram shows the distribution of R^2 values over monitor-specific performance (R^2) shown in (c) to monitor/location characteristics. Predictive power of each characteristic is calculated as the estimated change in monitor R^2 when each characteristic is increased from the 5th to 95th percentile of its distribution. Points show central estimates, and line segments show 95% confidence intervals. State outlines are reproduced with permission from the US Census Bureau TIGER/Line Shapefiles.⁵¹

Model Tuning and Validation. We aimed to predict smoke pollution for a 10 km grid over the conterminous US, so our measure of model performance needed to account for the fact that we would be predicting for out-of-sample locations and that those locations would have a median distance of about 50 km (population-weighted average of about 18 km) from EPA stations (Figure S9). Given concerns of information leakage between training and testing sets for proximate EPA stations, we used grid cells from the coarsest input (MERRA-2, 0.5° latitude \times 0.625° longitude, ~50 km) to define spatial folds, that is, the disjoint sets of training and testing stations used, respectively, for model training and model evaluation. Splitting train and test data sets by station rather than the more conventional method of random splitting by observation (in which a given station can contribute data to both train and test) is a more demanding prediction task^{34,61}—and using coarser spatial blocks further increases the difficulty of the prediction task⁶²—but is a more realistic test of how well the

model would do predicting time series of smoke $PM_{2.5}$ in a new location with no training data.

For model architecture, we fit gradient boosted trees,⁵⁸ which balance nicely our twin goals of model parsimony and high predictive power in related applications (e.g., Chen et al.,63 Zamani Joharestani et al.,64 and Xu et al.65). We performed fivefold nested spatial cross-validation, with hyperparameter tuning in the inner loop performed with fourfold cross-validation (Figure 1b; see the Supporting Information). All model tuning and training was performed on observations with smoke overhead. Based on preliminary tests, we used regression with the root mean squared error (RMSE) as the objective function, which performed better than pseudo-Huber loss and Tweedie regression—loss functions that, in principle, could respectively help with outliers in the data that the squared error is sensitive to and help with skewed non-negative distribution of outcomes. To measure variable importance, we use gain or the contribution to improvements in model performance from splits on each feature.

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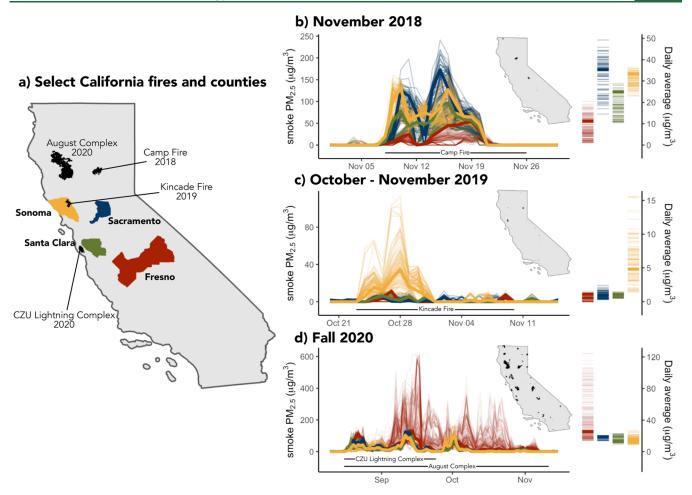


Figure 3. Predictions show remarkable spatial and temporal variation in smoke pollution from recent California fires. (a) Black shapes are select fires from November 2018, October to November 2019, and Fall 2020, and colored areas are four select California counties. (b–d) Daily smoke pollution varies between and within counties, and increases in smoke pollution temporally match with fire events. Thin lines show smoke pollution in grid cells within the county, while thick lines show population-weighted average for the county. Time ranges of select fires are shown along the horizontal axis. Inset maps show all fires burning during the time range of the panel. To the right, dashes show average smoke pollution over the time range in the grid cell (thin dashes) and county (thick line). Fire perimeters and time ranges are reproduced with permission from the CAL FIRE Fire Perimeter data set.⁷² State and county outlines are reproduced with permission from the US Census Bureau TIGER/Line Shapefiles.⁵¹

Assessing Model Performance. We measured model performance by comparing observed smoke PM2.5 with model predictions after truncating model predictions at zero. We calculate three metrics for evaluation: the overall R^2 , the "within" R^2 after netting out time-invariant differences in smoke PM25 across stations and changes in smoke over time common to all stations, and the RMSE on the held-out test set for each of the five folds. The "within" R^2 , which is calculated by regressing observed smoke PM2.5 on predicted smoke PM2.5 while including sets of station and year fixed effects (i.e., separate intercepts for each station and each year), is meant to measure performance in the context of a common downstream inference task in which within-location variation over time, rather than variation between locations, is used to isolate the impact of pollutants or other environmental exposures from other factors that might also shape human outcomes. To understand performance across days with any smoke, and across days with dense smoke, we calculate these metrics on the full sample of smoke days and the set of observations with smoke pollution of at least 50 μ g/m³.

RESULTS

Model Performance. Spatial out-of-sample model performance, measured over all observations using predictions from models trained on the other four folds, was an R^2 of 0.67, that is, our model explained two-thirds of the variation in smoke PM2.5 at stations not used in training. Our "within" performance $(R^2 = 0.65)$ approached our overall R^2 , indicating that our model is capably predicting local, temporal variation in smoke and not simply differences in average smoke across locations or between years. The model performed well over the entire range of observed smoke PM_{2.5} (Figure 2a and Table S4), including accurate prediction without saturation at very high daily PM_{2.5} levels, which has historically been a challenge for both numerical models and statistical models that often focus on PM_{2.5} observations below a certain threshold^{33,61,66} (although, see Reid et al.,³⁴ for an exception). We also compared model predictions to an alternative data set of ambient PM2.5 at private, non-reference-grade PurpleAir monitors⁶⁷ and found similar performance $(R^2 = 0.70)$ (Figure \$18); this additional data set was not used in training, so these results again demonstrate that our model performs well on completely new data and locations. Although the presence of

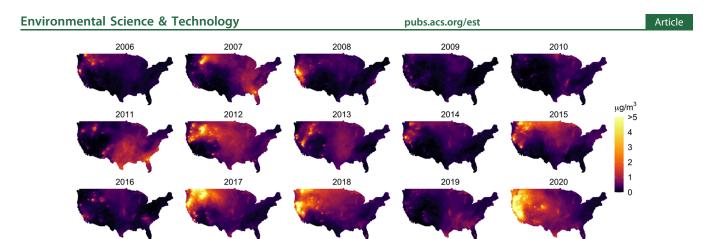


Figure 4. Daily average $PM_{2.5}$ from smoke by year. Each panel shows average daily smoke $PM_{2.5}$ in each location and each year, computed as the average over all days in each grid cell in each year.

satellite-based smoke plumes alone can be an imperfect indicator of surface-level pollution,⁶⁸ the performance of the model in predicting held-out surface $PM_{2.5}$ data suggests that the included features allow us to link changes in satellite measurements of aerosols and meteorological conditions to changes in surface $PM_{2.5}$. However, there remain a subset of smoke days where our model struggles to predict variation in surface $PM_{2.5}$, including days when a plume is overhead but observed smoke $PM_{2.5}$ values are near zero—likely cases when smoke is overhead but not mixing to the surface in a way that is predictable with our model features (Figures 2 and S18). However, among days with observed smoke $PM_{2.5} < 1 \ \mu g/m^3$, 94% of out-of-sample predictions are less than 5 $\mu g/m^3$.

Predicted AOD contributed most to model performance, followed by longitude, size of the nearest fire cluster, and dewpoint temperature (Figure 2b). To test the added predictive value from the two most computationally intensive feature sets, we also fit models without AOD predictions and without HYSPLIT features. Including AOD predictions had a higher positive impact on model performance (increase of 0.057 in R^2) than HYSPLIT features (increase of 0.014 in R^2 , Figure S10).

We calculated monitor-specific performance when each monitor was not used in model training. We found that the median station performance was comparable to overall model performance ($R^2 = 0.63$), with 90% of monitor-specific R^2 values falling between 0.21 and 0.88. Model performance varied spatially, with often very high performance at EPA monitors throughout California, the Pacific Northwest, the upper Midwest, and the Northeast and lower performance at monitors in the Southwest and the South (Figure 2c). To better understand this spatial variation in model performance, we then fit simple linear models between station-level R^2 and cross-sectional covariates, including new covariates from an alternative land cover classification (Figure S11).⁶⁹⁻⁷¹ We found that after latitude and longitude, shrubland land cover and average and variance in smoke pollution were most predictive of model performance at individual stations, as measured by the absolute predicted change in R^2 when a covariate is changed from its 5th to 95th percentile (Figure 2d). Model performance was higher in locations with higher average and variance in smoke PM2.5 and lower in locations with higher percentage shrubland land cover. Higher variance in non-smoke pollution was also associated with worse model performance. Shrubland areas may have less variation in smoke $PM_{2.5}$, more variation in non-smoke $PM_{2.5}$, leading to less precise estimates of smoke $PM_{2.5}$, or a different relationship between satellite AOD and ground $PM_{2.5}$ due to land cover, any of which could result in lower performance in smoke $PM_{2.5}$ predictions. These results help account for why performance was substantially higher in the Pacific Northwest (high average and variance in smoke $PM_{2.5}$, forest landcover) versus the desert Southwest (low average and variance in smoke $PM_{2.5}$, shrubland landcover); other efforts to predict total $PM_{2.5}$ have also performed substantially worse in the desert Southwest. 5,33,34 By examining individual station time series, we also found that our lowest performing stations were often characterized by a small number of extreme outliers in the ground measurements (Figure S12).

Patterns and Trends in Predicted Smoke $PM_{2.5}$. Predicted daily smoke $PM_{2.5}$ shows remarkable temporal and spatial variation around fire events, consistent with observed smoke behavior around individual fires (Figure S13). For example, Figure 3 shows model predictions for time periods surrounding three specific fire events⁷² in California over multiple years. Predicted smoke $PM_{2.5}$ showed increases corresponding to the onset of known fires, with variation in smoke $PM_{2.5}$ within fire events over time and across locations within counties (Figure 3b–d). These heterogeneities aggregate to marked differences in total smoke $PM_{2.5}$ concentrations between nearby locations and highlight the differences lost when using binary smoke measures or when interpolating between monitors.

Daily gridded estimates can also be aggregated to larger temporal or spatial scales to characterize broader patterns and trends in smoke concentrations. We measured the contribution of smoke to annual PM_{2.5} by aggregating daily predictions to the annual level across the contiguous US and found that some locations experienced over 2000 μ g/m³ of daily accumulated $\mathrm{PM}_{2.5}$ from smoke in the span of a year, equivalent to average annual PM_{2.5} levels 5 μ g/m³ higher (Figure 4). This level is equivalent to roughly half of the overall annual average $PM_{2.5}$ concentrations from all sources across much of the US. Annual hotspots of high smoke PM_{2.5} correspond to locations of large fires (Figures S15 and S16), although impacts often extend beyond the burned areas. The locations most affected varied by year but were primarily concentrated in the West, with larger areas affected in recent years, particularly 2017, 2018, and 2020. The Southwest and Northeast experienced the least smoke pollution over the study period.

b) Annual smoke PM_{2.5} by state

a) Decadal change in smoke PM_{2.5}

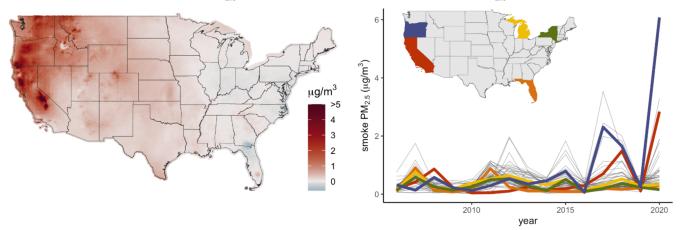


Figure 5. Over the last decade, smoke $PM_{2.5}$ has increased in much of the US, particularly in Western states, but some areas in the South and East have seen modest declines. (a) Decadal change in smoke $PM_{2.5}$ is the difference in daily average smoke $PM_{2.5}$ during 2006–2010 and 2016–2020. (b) Population-weighted average smoke $PM_{2.5}$ shows marked increases in Western states and little change in Eastern states. Each line indicates a state, with the inset map showing highlighted states whose colors match the lines corresponding to the states in the main panel. State outlines are reproduced with permission from the US Census Bureau TIGER/Line Shapefiles.⁵¹

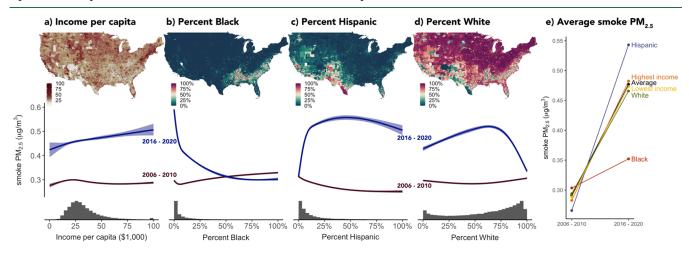


Figure 6. One decade ago, smoke $PM_{2.5}$ levels did not differ meaningfully by income, race, or ethnicity, but recent exposure patterns now differ by race, ethnicity, and, to a smaller extent, income. (a) All income levels experienced similar smoke $PM_{2.5}$ concentrations from 2006 to 2010 and increases in smoke $PM_{2.5}$ to recent years (2016–2020), with the greatest increases occurring at the highest income levels. (b) Smoke $PM_{2.5}$ concentrations have fallen in heavily non-Hispanic Black tracts, consistent with the modest declines seen in the Southeastern region. (c) Concentrations have increased most in tracts with Hispanic populations, corresponding to higher percent Hispanic in Western and Southwestern regions. (d) Smoke $PM_{2.5}$ increases were the smallest in tracts that were heavily non-Hispanic white, concordant with the more modest smoke $PM_{2.5}$ increases in the Midwest and Northeast. For all panels, lines are spline fits between income/race/ethnicity and average smoke concentrations in 2006–2010 (purple) and 2016–2020 (blue) in US Census tracts, with shaded areas showing bootstrapped 95% confidence intervals, while histograms show the distribution across tracts. Tract-level smoke $PM_{2.5}$ is an area-weighted average of smoke $PM_{2.5}$ in intersecting grid cells. Income per capita is top-coded to \$100,000. (e) We also calculate the average annual smoke $PM_{2.5}$ concentrations for the average Hispanic individual (blue), White individual (green), Black individual (red), and US resident (black) and individuals living in census tracts in the highest (orange) and lowest (yellow) income quintiles. Census tract outlines are reproduced with permission from the US Census Bureau TIGER/Line Shapefiles.⁵¹

To better understand longer-run changes in smoke $PM_{2.5}$ concentrations, we calculated the decadal change in smoke pollution as the difference in average annual smoke $PM_{2.5}$ from 2006–2010 to 2016–2020, with 5 year windows used to avoid undue influence from extreme years, such as 2020. Locations west of the Mississippi River saw increases in smoke pollution in the last decade, with the greatest increases in Washington, Oregon, and California (Figure 5a). Some of these Western regions saw decadal increases in an annual smoke $PM_{2.5}$ of 5 $\mu g/m^3$ or greater, an amount comparable in absolute magnitude to the reduction in $PM_{2.5}$ brought about by the

Clean Air Act in the US.^{74,75} Some locations in the Southeast and Northeast saw modest (<1 μ g/m³) declines in smoke PM_{2.5}, consistent with a decline in burned area observed in existing fire products in many states in the Southeast, East, and Midwest (i.e., Monitoring Trends in Burn Severity,⁷⁶ Figure S14) and a decline in days with heavy smoke plumes in the air in the Southeast.³¹ The notable declines on the Georgia-Florida border and in central Idaho may be due to the 2007 fires in those areas, which were associated with elevated annual smoke PM_{2.5} (Figures 4 and S16). We also quantify population-weighted concentrations by state for each year

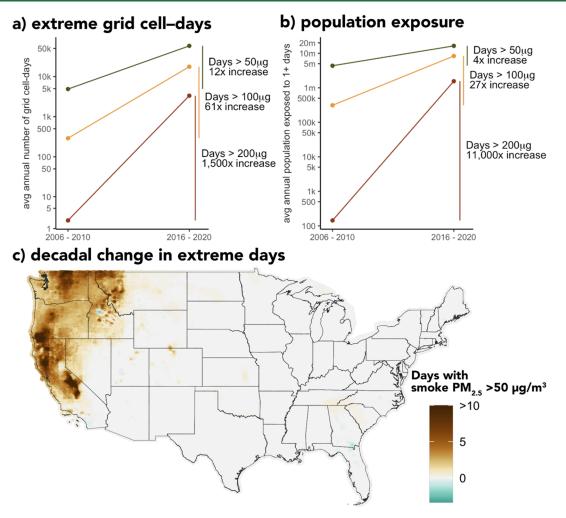


Figure 7. Changing occurrence of smoke $PM_{2.5}$ extremes. (a) Decadal change in the number of grid cell days with smoke $PM_{2.5}$ above 50 (green), 100 (orange), and 200 μ g/m³ (red), computed as the difference in average annual counts during 2006–2010 and during 2016–2020. Annotations show the change in multiples of the baseline rate. (b) Decadal change in the population in the US living in places with at least 1 day per year of smoke above 50, 100, or 200 μ g/m³. For instance, over 16 million people on average in the US lived in places with at least 1 day above 50 μ g/m³ per year in the last 5 years, a fourfold increase from a decade ago. (c) Map of decadal change in extreme days with smoke $PM_{2.5}$ over 50 μ g/m³. State outlines are reproduced with permission from the US Census Bureau TIGER/Line Shapefiles.⁵¹

and find that concentrations were worst for the average Oregon resident in 2020, with annual smoke $PM_{2.5}$ levels over 50 times higher than a decade previous (Figure 5b). On the other hand, residents in Midwestern, Northeastern, and Southern states had similar levels of smoke $PM_{2.5}$ over the last 15 years.

Differences in Smoke Pollution by Race, Ethnicity, and Income. Although PM2.5 from smoke is growing throughout most of the country, the geographic patterns in smoke PM_{2.5} may result in differential exposure for different populations. Understanding the magnitudes and sources of disparities in environmental exposures across income and racial/ethnic groups is a key question in environmental justice. We merge our gridded data with census tract level data on income, race, and ethnicity from the US Census Bureau American Community Survey (ACS)⁷⁷ to quantify the levels and trends in ambient concentrations. From 2006 to 2010, average annual smoke PM2.5 did not differ meaningfully by income or by the percent of tract population represented by non-Hispanic black, non-Hispanic white, or Hispanic individuals (Figure 6). Over the last decade, smoke $PM_{2.5}$ concentrations have grown fastest for predominantly Hispanic

and non-Hispanic white populations, while predominantly non-Hispanic black census tracts have seen a slight decline in smoke $PM_{2.5}$ (Figure 6). Across all income levels, smoke $PM_{2.5}$ has increased, but that increase has been slightly larger in higher income census tracts. These changes reflect the broad geographic patterns in smoke $PM_{2.5}$ trends (Figure 5), as well as the geographic concentrations of different racial and ethnic groups in the country. We also compare average annual smoke $PM_{2.5}$ levels to gross domestic product (GDP) per capita by county⁷⁸ and to population density per grid cell.⁷⁹ We find that smoke $PM_{2.5}$ concentrations have increased more in counties with higher GDP per capita and have been the smallest in intermediate population density areas, larger in high population density areas, and the largest in very low density areas (Figure S17).

Changing Occurrence of Daily Extremes. Finally, the primary goal of our analysis was to ensure that our model had low bias in predicting not only moderate (and more common) daily smoke $PM_{2.5}$ concentrations but also extreme levels of daily smoke $PM_{2.5}$. Quantifying exposures to extremes is critical for understanding health and related impacts, both because impacts might be non-linear in exposure and because

compensatory behavior (e.g., staying indoors) differs substantially by the level of exposure.³¹ Whether resulting concentration—response functions are concave, convex, or linear is not well established in the literature⁴ but has important implications for the magnitude of societal impacts and the optimal policy response.

We estimate a substantial increase in exposure to extremes in daily smoke PM_{2.5} over the last decade. Using thresholds of 50, 100, or 200 μ g/m³ of daily smoke PM_{2.5} concentrations to denote extremes, we calculate a remarkable 12-1500× decadal increase in the annual average count of grid cell days with concentrations above these thresholds (Figure 7a). Because many of these grid cells might be in relatively unpopulated areas of the Western US, we also calculate the number of people residing in locations where at least a day per year exceeded these thresholds, using population estimates from WorldPop.⁷⁹ We calculate that each year, on average, 16.4 million people in the US lived in places with at least 1 day of smoke PM_{2.5} above 50 μ g/m³ during the 2016–2020 period, a fourfold increase from the 2006 to 2010 average (Figure 7b). While less than half a million people lived in areas experiencing a day per year of smoke $PM_{2.5}$ concentrations over 100 μ g/m³ a decade ago, this number increased to over 8 million in recent years. In proportional terms, increases were the largest for the most extreme threshold, with almost nobody experiencing a day per year above 200 μ g/m³ a decade ago, but now, over 1.5 million people are residing in locations routinely exposed to these conditions. The locations experiencing the greatest increase in days with extreme smoke PM2.5 are concentrated in the Western US, particularly Washington, Oregon, California, Idaho, Montana, and Nevada (Figure 7c).

DISCUSSION

Here, we produce 10 km resolution estimates of smoke $PM_{2.5}$ over the contiguous US from 2006 to 2020 on a daily basis. These predictions perform well over the range of observed smoke $PM_{2.5}$ and vary predictably in performance by region (Figure 2). These predictions capture small-scale variation in smoke $PM_{2.5}$ within counties and within fire events (Figure 3) and larger scale interannual variation in smoke pollution (Figure 4). We find steep increases in average smoke pollution and days with smoke $PM_{2.5}$ extremes over the last decade, particularly in the Western US (Figures 5 and 7), resulting in a changing profile of smoke $PM_{2.5}$ concentrations (Figure 6). Consistent with recent work,^{4–6} these predictions suggest that the increases in $PM_{2.5}$ from wildfire smoke are rapidly undoing much of the improvements in air quality from the last few decades, at least throughout many areas of the Western US.

In comparison to many existing efforts to estimate smoke $PM_{2.5}$ using CTMs with and without emissions from fires or through sensitivity analyses embedded in CTMs,⁸⁰ our approach uses statistical, machine-learning models fit to inferred smoke $PM_{2.5}$ at EPA monitors. Although CTM-based smoke $PM_{2.5}$ estimates trade-off between the computational cost, spatial coverage, and spatial resolution, our statistical approach can achieve high resolution (10 km) estimates over the entire contiguous US, can be validated against long time series of monitor observations, and can easily scale to large areas due to the fast prediction speeds after models have been trained. On the other hand, CTM-based estimates can characterize subdaily variation in smoke $PM_{2.5}$, while our estimates are currently limited to daily resolution. Finally, CTMs can provide speciated pollutant estimates of

 $PM_{2.5}$, albeit with substantial uncertainty in concentration estimates,²² while our focus is on total smoke $PM_{2.5}$.

Our work is related to a recent interpolation-based approach used to estimate wildfire-attributable PM_{2.5} across the US.⁵ While we take a similar approach to measuring smoke PM_{2.5} in ground data, we diverge in using a range of satellite- and model-based inputs, rather than interpolation, to estimate smoke PM_{2.5} in locations without stations. Because interpolation by design smooths the input data, such an approach could understate extreme concentrations, both in locations without monitors and on the large number of smoke days when specific monitors do not report concentrations. Finally, we validate our approach against 15 year time series of heldout station observations and against independent private sensor data, which stands in contrast to the single-season evaluation in earlier work.5 Our work complements recent machine learning-based efforts to use satellites and other inputs to predict total PM_{2.5} concentrations over time.^{33,34}

As with most available data sets on specific pollutants, variation in estimated smoke $PM_{2.5}$ in our data is likely correlated with variation in other pollutants including NO₂ and ozone, ^{47,81} although the emission, formation, and evolution of those other pollutants are an evolving area of research.^{82,83} Downstream applications that wish to use our data to isolate the impact of $PM_{2.5}$, specifically on outcomes, must then take care to account for co-varying pollutants that could also affect outcomes. Such applications should also consider accounting for other covarying meteorological variables such as temperature and precipitation, although how best to account for the role of these variables is not necessarily clear, as these variables are plausibly both a driver of wildfire activity (and thus not a confound) and an independent source of impact (and thus possibly a confound).

Future advancements to smoke PM2.5 estimates could include improvements to both the monitor-based training data and to our machine learning-based smoke prediction pipeline. Precision of smoke PM2.5 estimates at monitors-our training data-is a function of the precision of the non-smoke counterfactual, that is, the estimated background rate of PM_{2.5} absent smoke. We currently use median PM2.5 from days without smoke, which accounts for station-specific differences, seasonal variation within the station, and trends over time in PM_{2.5} levels but does not account for more granular variation in other time-varying non-smoke sources of PM_{2.5}. This may result in imprecision in our training smoke PM_{2.5} estimates, as anomalous PM2.5 may be a noisy estimate of the concentrations from smoke, especially in locations with high variation in background non-smoke PM2.5 which was associated with poorer model performance (Figure 2). Future efforts could more explicitly model background non-smoke PM2.5, perhaps using machine learning and other information on time variation in other PM2.5 sources. Future model development might also benefit from the increasing availability of monitor data as large numbers of private sensors, such as PurpleAir monitors, come online (Figure S18). In this study, we use these latter data as a source of independent evaluation, but such data could also be useful in model training, although their use should consider that these monitors are neither reference grade nor regularly calibrated. Several procedures have been developed for improving PM_{2.5} estimates derived from PurpleAir monitors.⁸⁴⁻⁸⁸ While the most appropriate calibration likely depends on the setting, we utilized the Barkjohn correction⁸⁴ because we found in previous work³¹ that it

performed best at predicting out-of-sample PM2.5 at EPA reference grade monitors. As higher spatial and temporal densities of training data were predictive of increased model performance (Figure 2d), future inclusion of these data sources should improve the accuracy of estimates. Other features and data sources such as smoke plume height, spatial lags of meteorology, and indicators of atmospheric mixing such as air temperature at different vertical heights have been found in other settings to improve total PM2.5 estimates, predict variation in the relationship between PM_{2.5} and AOD, or have the potential to improve the model's ability to identify when smoke mixes to the surface, something the current model occasionally struggles with, as evidenced by the range of predicted values on days with very low observed smoke PM2.5 values.^{89–92} Future advances could also include alternative machine learning models, such as convolutional neural networks, that take advantage of the spatial information instead of features at a single point and have been found to provide good performance on total PM2.5.93 While our estimates rely on plume boundaries drawn by NOAA analysts over the contiguous US, automation of plume identification-a task for which early computer vision work has shown promise^{94–96}—could allow for generalization of this approach to other geographic regions, an effort of increasing importance as wildfires grow in many parts of the world.^{11,97-99} Finally, uncertainty quantification from machine learning models is an active area of research, and future improvements to these estimates could include more granular quantification of uncertainty.

A growing literature estimates the impact of smoke on a range of outcomes including health outcomes such as mortality, hospitalizations, and birth outcomes, 100,101 economic outcomes including labor productivity and economic output,¹⁰² and cognitive outcomes such as test scores and mental health.⁹⁴ Many of these studies have been limited to short time spans, small geographic ranges, or binary measures of smoke concentrations, in part due to lack of broad-scale validated data on pollutant concentrations attributable to wildfire smoke.¹⁰³ Our new estimates should enable improvements in the breadth and precision of these studies and help refine our understanding of how growing smoke PM2.5 concentrations affect a range of societal outcomes and are well suited for studying the impact of more common, low-level smoke concentrations and increasingly frequent periods of extreme concentrations. Our model's strong ability to predict within-location variation in smoke PM2.5 over time and to generate long time series of concentration estimates are particularly beneficial for understanding societal impacts, given the frequent reliance of modern causal inference approaches on temporal rather than spatial variation in estimating the impact of environmental exposures on societal outcomes. Our estimates can thus help establish smoke-specific concentration-response functions across a range of societal responses of interest and, by more accurately measuring extreme exposures, help better characterize potential non-linearities in these functions. Finally, smoke PM_{2.5} can also be linked to antecedent climate conditions to better understand the past local and regional influence of climate on smoke PM25 concentrations and to inform how smoke pollution might evolve under future climate.

DATA AVAILABILITY

Data and code to replicate all results in the main text and supplementary materials are available at https://github.com/echolab-stanford/daily-10km-smokePM.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c02934.

Additional model details including model tuning and model inputs, assessment of smoke plume accuracy and smoke $PM_{2.5}$ estimates using speciated data, additional model performance comparisons, and trends in and annual burned areas (PDF)

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Author Contributions

All authors contributed to the conception and design of the study. M.L.C., J.L., J.W., S.H.-N., and A.D. contributed to data extraction and processing. M.L.C. conducted AOD prediction, and J.L. conducted HYSPLIT simulations. M.L.C. and J.L. performed model tuning and evaluation. M.L.C. led the analyses of model performance, smoke PM_{2.5} patterns, and differential exposures, and M.B. led the analysis of daily

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extremes. All authors contributed to analyzing results and writing the paper.

Notes

The authors declare no competing financial interest.

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